Calculation of Stark-induced absorption on the $6s6p\,^3P_1 - 6s^2\,^1S_0$ transition in Hg

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We carry out relativistic many-body calculations of the Stark-induced absorption coefficient on the 254-nm $6s6p\,^3P_1(F=1/2) - 6s^2\,^1S_0$ line of ^{199}Hg atom, the effect considered before by Lamoreaux and Fortson using a simple central field estimate [Phys. Rev. A **46**, 7053 (1992)]. The Stark-induced admixing of states of opposite parity opens additional M1 and E2 transition channels. We find that the resulting M1-E1 absorption dominates over E2-E1 absorption. The value of the E2-E1 absorption coefficient depends strongly on the details of treatment of the correlation problem. As a result, our numerical values differ substantially from those of the earlier central field calculation. Reliable calculation of this effect can enable a useful experimental check on the optical technique being used to search for a permanent electric dipole moment of the ^{199}Hg atom.

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I. INTRODUCTION

The $F = 1/2 \to F = 1/2$ electromagnetic transition between two atomic states of opposite parity has necessarily the electric-dipole (E1) character. However, an application of the external E-field $\mathcal{E}_s$ breaks the spherical and mirror symmetries of the atomic Hamiltonian and opens all multipolar transition channels. To the lowest order in $\mathcal{E}_s$, the transitions are determined by the M1 (magnetic-dipole) and E2 (electric-quadrupole) channels. These effects modify the absorption coefficient of the atomic sample, the corrective M1 and E2 terms scaling linearly with the electric field [1, 2].

Lamoreaux and Fortson [2] have focused on a specific setup, relevant for the search of the permanent electric dipole moment (EDM) of Hg atom [3, 4] (non-vanishing EDM would violate P- and T-reversal symmetries and be a clear signature of new physics beyond the standard model of elementary particles). They considered exciting the 254-nm $6s^2\,^1S_0 \to 6s6p\,^3P_1$ transition of ^{199}Hg atom. This isotope has the nuclear spin $I = 1/2$. A laser resolves the hyperfine structure of the $^3P_1$ level. It resonantly drives transitions from a given magnetic M_{F_i} sublevel of the $F_i = 1/2$ ground state to the $F_f = 1/2$ level of the excited state. Then, for the $F_i = 1/2 \to F_f = 1/2$ transitions, the relative change in the absorption coefficient α may be parameterized as [2]

$$\frac{\delta\alpha}{\alpha} = (a_{M1} + a_{E2})\ \ (\hat{\varepsilon}_L \cdot \mathcal{E}_s)\left(\hat{\mathbf{k}}_L\ \times\ \hat{\varepsilon}_L\right)\cdot\left(\frac{\langle\mathbf{F}_i\rangle}{F_i}\right)\ . \quad (1)$$

Here $\hat{\varepsilon}_L$ is the polarization vector and $\hat{\mathbf{k}}_L$ is the direction of propagation of the laser wave. $\langle\mathbf{F}_i\rangle$ is the expectation value of the total angular momentum, i.e., the nuclear polarization in the ground $J_i = 0$ atomic state.

In the current ^{199}Hg EDM experiment [4], the 254-nm transition is used to monitor the nuclear spin direction and thereby detect EDM-induced shifts in nuclear spin precession, which will be linear in an external electric field. The Stark interference effect on the 254 nm absorption given in Eq. (1) also is linear in electric field $\mathcal{E}_s$ and depends upon the nuclear spin direction. A reliable calculation of this effect can enable a useful check on the EDM method when the effect is measured under the same experimental conditions as in the EDM search [5].

II. EXPRESSIONS FOR ABSORPTION COEFFICIENTS

The goal of this paper is to compute the atomic-structure coefficients a_{M1} and a_{E2}. One may qualitatively understand the appearance of M1 and E2 admixtures in Eq. (1) as follows. The Stark-induced transition amplitude in a laser field is composed from terms linear in the interactions with the external E-field, $-\mathbf{D}\cdot\mathcal{E}_s$ ($\mathbf{D}$ being the dipole operator), and the driving 2^k-pole laser field. We may recouple the products of the two tensors ($\mathbf{D}$ and the laser EM multipolar interaction); the resulting compound operators have the multipolarities of $|k-1|, k, k+1$. For the $F_i = 1/2 \to F_f = 1/2$ transition, k would be limited to 1 and 2. The additional constraint imposed by the parity selection rule yields the M1 and E2 multipolar couplings.

We derived the expressions for a_{M1} and a_{E2} using the multipolar expansion of the plane EM wave and the first-order perturbation theory in the Stark field for the wavefunction. We employ a geometry where the quantization axis $\hat{z}$ is chosen along the k-vector of the linearly-polarized laser. The DC Stark-field and the laser polarization are aligned along the x-axis, and the atom has a definite value of $\hat{F}_y$ in the initial state. This particular choice of geometry is convenient for working with the most general relativistic expressions for the multipolar transition operators [6]. By evaluating Eq. (1) of Ref. [2] in this geometry, we identify the following expressions for

the structure factors

$$a_{M1} = \sqrt{\frac{2}{3}}\frac{R^{M1}}{\langle n_i J_i||\mathbf{r}||n_f J_f\rangle}, \quad (2)$$

$$a_{E2} = -\frac{1}{4}\sqrt{\frac{3}{5}}k_L\frac{R^{E2}}{\langle n_i J_i||\mathbf{r}||n_f J_f\rangle}. \quad (3)$$

Unless specified otherwise, here and below we use the atomic units, $\hbar = |e| = m_e = 1$, and the Gaussian units for EM equations. Here $\langle n_i J_i||\mathbf{r}||n_f J_f\rangle$ is (within a factor of -1 corresponding to the charge of the electron) the conventional reduced dipole matrix element for the $6s^2\,{}^1S_0 \to 6s6p\,{}^3P_1$ transition and $k_L = \omega_L/c$ is the magnitude of the wavevector of the laser. The quantities R^{M1} and R^{E2} are sums over a complete set of intermediate states; these sums arise due to the Stark-induced perturbation and involve the static E1 operator and multipolar AC couplings to the driving laser field. Each of the sums, R^{M1} and R^{E2}, may be further split into two sums $S^{M1/E2}_{i/f}$, subscript i or f indicating which of the states, initial or final, is Stark-perturbed,

$$R^{M1} = -S^{M1}_i\,(1^o) + S^{M1}_f\,(1^e)\,, \quad (4)$$

$$R^{E2} = -\frac{2}{3}\sqrt{2}S^{E2}_i\,(1^o) - 2\sqrt{\frac{2}{15}}S^{E2}_f\,(2^e)\,. \quad (5)$$

The argument of the sums $S^{M1/E2}_{i/f}(J^\pi)$ indicates the total angular momentum J and the parity π of the intermediate states as fixed by selection rules. Explicitly, the reduced sums are

$$S^{M1}_i\,(J^\pi_n = 1^o) = \sum_n \frac{\langle n_i J_i||\mathbf{r}||n_n J_n\rangle\langle n_n J_n||Q^{(M1)}||n_f J_f\rangle}{E_i - E_n},$$

$$S^{M1}_f\,(J^\pi_n = 1^e) = \sum_n \frac{\langle n_i J_i||Q^{(M1)}||n_n J_n\rangle\langle n_n J_n||\mathbf{r}||n_f J_f\rangle}{E_f - E_n},$$

$$S^{E2}_i\,(J^\pi_n = 1^o) = \sum_n \frac{\langle n_i J_i||\mathbf{r}||n_n J_n\rangle\langle n_n J_n||Q^{(E2)}||n_f J_f\rangle}{E_i - E_n},$$

$$S^{E2}_f\,(J^\pi_n = 2^e) = \sum_n \frac{\langle n_i J_i||Q^{(E2)}||n_n J_n\rangle\langle n_n J_n||\mathbf{r}||n_f J_f\rangle}{E_f - E_n}.$$

We employ the relativistic formalism for the multipolar transition operators $Q^{(M1/E2)}$. Specific single-particle reduced matrix elements computed using Dirac orbital parameterization of Ref. [6] are

$$\langle i||Q^{(EJ)}||j\rangle = \langle\kappa_i||C_J||\kappa_j\rangle \times \int_0^\infty r^J\{G_i(r)G_j(r) + F_i(r)F_j(r)\}dr, \quad (6)$$

$$\langle i||Q^{(MJ)}||j\rangle = \frac{\kappa_i + \kappa_j}{J+1}\langle -\kappa_i||C_J||\kappa_j\rangle \times \int_0^\infty r^J\{G_i(r)F_j(r) + F_i(r)G_j(r)\}dr. \quad (7)$$

In both expressions we used the long-wavelength approximation, as $\alpha\omega_L \ll 1$. In these expressions, $G(r)$ $(F(r))$ are the large (small) radial components of the Dirac bispinor, κ are the relativistic angular quantum numbers, and $C_J(\hat{r})$ are the normalized spherical harmonics.

III. ATOMIC-STRUCTURE FORMALISM

Mercury atom has two valence electrons outside a closed-shell core and we start our calculations with the so-called frozen core (V^{N-2}) Dirac-Hartree-Fock (DHF) approximation. In this approximation, the core orbitals are obtained self-consistently, while excited (valence) orbitals are subsequently generated by solving the Dirac equation in the resulting potential of the core. Such orbitals correspond to the Hg^+ valence orbitals. They are used as a basis for the standard configuration interaction (CI) technique for two valence electrons (see, e.g. [7]). We refer to this approximation as CI-DHF. Further significant improvement of the accuracy of the calculations is achieved when the standard CI technique is combined with many-body perturbation theory (MBPT) to include correlations of the valence electrons with the atomic core (CI+MBPT).

The CI+MBPT formalism has been discussed in a number of papers (see, e.g., [8, 9, 10]). The effective operator (self-energy, $\hat{\Sigma}$) arising from the core polarization may be split into a single-particle, $\hat{\Sigma}_1$, and a two-particle, $\hat{\Sigma}_2$, part acting in the model space. Qualitatively, a field of the valence electron induces an electric dipole of the polarizable core: $\hat{\Sigma}_1$ describes an interaction of the valence electron with the self-induced core dipole, while $\hat{\Sigma}_2$ describes its interaction with the core dipole induced by the *other* valence electron. We compute the self-energy correction in the second order of MBPT for the residual Coulomb interaction. Effects of higher orders will be also included in a semi-empirical fashion, discussed below.

We use the Brillouin-Wigner flavor of MBPT [7] to avoid the "intruder-state problem", when the virtual core excitations inside $\hat{\Sigma}_2$ become resonant with the states of the valence subspace. Finally, we emphasize that our computations are *ab initio* relativistic and employ the Dirac equation and bi-spinors throughout the entire calculation.

We use the second-order MBPT to calculate the self-energy operators $\hat{\Sigma}_1$ and $\hat{\Sigma}_2$ via direct summation over a complete set of single-electron states. This set of basis states is constructed using the B-spline technique [11]. We use 40 B-splines of order 9 in a cavity of 40 Bohr radius. The same basis of the single-electron states is also used in constructing the two-electron basis states for the CI calculations. We employ partial waves $\ell = 0 - 4$ and the 14 lowest states above the core in each partial wave ($s_{1/2}$, $p_{1/2}$, $p_{3/2}$, etc.) for the valence CI subspace and $\ell = 0 - 5$ and 30 lowest states in each partial wave for internal summations inside the self-energy operator.

Higher-order correlations are also included in $\hat{\Sigma}$ in a way similar to Ref. [12]. The $\hat{\Sigma}_1$ operator depends on the symmetry of the valence orbital. Therefore, we have a set

of different $\hat{\Sigma}_1$ operators for $s_{1/2}$, $p_{1/2}$, $p_{3/2}$, etc. states. An analysis of the spectra of Hg (see Table I) shows that accurate treatment of $\hat{\Sigma}_1$ is most important for s-electrons, because the ground $6s^2$ state and other states with s electrons come close to the core and therefore core-valence correlations must be sizeable for them. In contrast, the core-valence correlations are much smaller for more diffuse p and d orbitals. It turns out that the best accuracy is achieved if the all-order $\hat{\Sigma}_1^{\infty}$ [13] operator is used for the s electrons, the second-order $\hat{\Sigma}_1$ is employed for the p electrons and no $\hat{\Sigma}_1$ is included for d and higher waves.

Higher-order contributions to $\hat{\Sigma}_2$ are included semi-empirically via screening factors which modify Coulomb integrals of the second-order $\hat{\Sigma}_2$ (see Ref. [12] for details). The values of these factors are $f_0 = 0.9$, $f_1 = 0.72$, $f_2 = 0.98$, $f_3 = 1$, $f_4 = 1.02$ and $f_5 = 1.02$. These values are found from comparing second-order and all-order $\hat{\Sigma}_1$.

Finally, we further rescale the $\hat{\Sigma}_1$ operator for the s and p electrons to fit the experimental spectrum better. The rescaling coefficients are $\lambda_s = 1.0961$ and $\lambda_p = 0.8675$. We use the same λ_p for $p_{1/2}$ and $p_{3/2}$ waves. Note that $\lambda_s > 1$ because high-order effects, included in $\hat{\Sigma}_1$ for the s electrons, significantly reduce its value. On the other hand, $\lambda_p < 1$ because the second-order MBPT always overestimates the correlation correction.

The resulting energies are listed and compared with experiment in Table I. A typical deviation from the experimental values is in the order of 100 cm^{-1}. Even after the scaling, the disagreement remains, as the number of fitting parameters is limited.

TABLE I: Experimental and theoretical energy levels of Hg (in cm^{-1}).

State	J	Exp. [14]	Theory
$6s^2$ ^{1}S	0	0.000	−13.79
$6s6p$ ^{3}P^o	0	37645.080	37458.26
	1	39412.300	39312.86
	2	44042.977	44265.45
$6s6p$ ^{1}P^o	1	54068.781	54180.72
$6s7s$ ^{3}S	1	62350.456	62171.92
$6s7s$ ^{1}S	0	63928.243	63672.24
$6s7p$ ^{3}P^o	0	69516.66	69211.87
	1	69661.89	69385.18
	2	71207.51	70094.95
$6s7p$ ^{1}P^o	1	71295.15	71189.34
$6s6d$ ^{1}D	2	71333.182	71295.01
$6s6d$ ^{3}D	2	71396.220	71353.26

The diagonalization of the CI+MBPT Hamiltonian provides us with the atomic wavefunctions and energies. While the wavefunctions already have correlation corrections built in, evaluating matrix elements requires additional inclusion of the so-called screening effect. This effect arises already in the first order in the residual Coulomb interaction and describes a re-adjustment of the core orbitals in response to an externally applied field. We incorporate the screening in the framework of the all-order many-body technique, the random-phase approximation (RPA). The RPA formalism (see, e.g., Ref. [15]) describes a linearized response to an oscillating perturbation. In this regard, while evaluating the reduced sums, we need to fix the driving RPA frequency for the entire set of matrix elements $Q^{(M1)}$ and $Q^{(E2)}$ at the photon frequency, $\omega_L = E_f - E_i$. However, for the dipole matrix elements (Stark mixing), the RPA frequency $\omega = 0$.

The evaluation of the sums S requires summing over a complete set of intermediate atomic states $|n_n J_n\rangle$. We use two approaches: (i) direct summation over states (this implies explicit computation of the atomic states and evaluation of matrix elements), and (ii) the Dalgarno-Lewis method. In the Dalgarno-Lewis method [16], the summation is reduced to solving the inhomogeneous Schrödinger (Dirac) equation (setup is similar to Ref. [17]). As an illustration, consider evaluation of the sum S_f^{M1}. It may be represented as

$$S_f^{M1} = \langle n_i J_i || Q^{(M1)} || \delta\Psi_f \rangle,$$

where $|\delta\Psi_f\rangle$ lumps corrections to the atomic wave function of the final state due to the external field. It satisfies an inhomogeneous equation

$$(\hat{H}_{\rm eff} - E_f)|\delta\Psi_f\rangle = -\mathbf{r}|n_f J_f\rangle, \qquad (8)$$

where $\hat{H}_{\rm eff}$ is the effective CI+MBPT Hamiltonian of the atom.

IV. NUMERICAL RESULTS

As an illustration of the CI+MBPT methodology, we start with calculations of the E1 matrix element and energy interval for the $6s^2\,{}^1S_0 - 6s6p\,{}^3P_1$ transition. This matrix element normalizes the Stark-induced corrections to the absorption coefficient, Eq. (1). The theoretical results at various levels of approximation and a comparison with the experimental values are presented in Table II. We observe that the core-polarization ($\hat{\Sigma}_1$) has a substantial effect on the energy interval, leading to an improvement in the theory-experiment agreement. While the CI+$\hat{\Sigma}_1$ value of the matrix element perfectly agrees with the experiment [18], such an agreement is fortuitous: including the screening correction to the Hamiltonian ($\hat{\Sigma}_2$) increases its value by a factor of 1.6; only the additional inclusion of the RPA screening and semi-empirical scaling moves the theoretical value into a 10% agreement with a 2%-accurate experiment. We find such an accuracy acceptable, as *ab initio* matrix elements of the intercombination (spin-forbidden) transitions are known [19] to be very sensitive to many-body corrections, the entire values being accumulated due to the relativistic effects. On the other hand, the matrix elements of *spin-allowed* transitions are stable with respect to inclusion of the MBPT effects (see, e.g., Ref. [19]). We will return to the evaluation of the accuracy of our calculations later.

TABLE II: Energy interval ΔE (in cm^{-1}) and the reduced electric-dipole matrix element (R, a.u.) for the $6s^2\,{}^1S_0 - 6s6p\,{}^3P_1$ transition in Hg atom in various approximations.

Approximation	ΔE	R
CI-DHF	31028	0.405
CI+Σ_1	37441	0.453
CI+Σ_1+Σ_2	37623	0.716
CI+Σ_1+Σ_2+RPA		0.577
as above but with all-order Σ_2	36947	0.512
as above but with scaled Σ_1 (Final)	39313	0.503
Experiment [14, 18]	39412	0.453(8)

The Stark-induced correction to the absorption coefficient involves two channels, M1 and E2. We start by discussing the more involved a_{E2} calculations. We need to compute two sums, $S_i^{E2}\,(J_n^\pi = 1^o)$ and $S_f^{E2}\,(J_n^\pi = 2^e)$. We carry out calculations (i) by direct summation over the 10 lowest-energy intermediate states of each symmetry (1^o and 2^e) and (ii) by using the Dalgarno-Lewis method. The latter method is equivalent to summing over infinitely many intermediate states. Both calculations use the most sophisticated CI+MBPT approximation (i.e., CI+Σ_1+Σ_2+RPA with semi-empirical scaling). The results are presented in Table III. An examination of contributions reveals that there are substantial cancellations inside individual sums. This leads to an enhanced sensitivity to correlations. For example, consider the value of the $S_i^{E2}\,(J_n^\pi = 1^o)$ sum truncated at the 10 lowest-energy levels. It changes from -35.6 to -28.23 (Table III) while progressing from the CI+Σ_1+RPA to the full CI+MBPT treatment. Additional cancellations occur when the reduced sums are combined into the quantity $R^{E2} = -\frac{2}{3}\sqrt{2}S_i^{E2}\,(1^o) - 2\sqrt{\frac{2}{15}}S_f^{E2}\,(2^e) \approx 24.13 - 32.84 = -8.71$. Notice that this value is several times smaller than the properly rescaled value of the largest contribution in Table III. These cancellations may lead to a poor accuracy of our resulting absorption coefficient

$$a_{E2} = -4.39 \times 10^{-3}\ \text{a.u.} = -0.0853 \times 10^{-8}/(\mathrm{kV/cm})\,.$$

This result was obtained using the *ab initio* matrix element from Table II. Notice that there is a phase ambiguity originating from atomic wavefunctions for sums S and the normalizing dipole matrix element. However, when these quantities are combined in Eq. (2,3), the ambiguous phase factors cancel out. In our particular computation, the sign of the dipole matrix element $\langle 6s^2\,{}^1S_0||\mathbf{r}||6s6p\,{}^3P_1\rangle$ is fixed by the first entry of Table III.

We proceed to a comparison with results of Ref. [2]. These authors use a simplified approach in which a true many-electron problem is reduced to a set of single-electron problems. For the E2 interference they use the Dalgarno-Lewis summation method based on the DHF orbitals of the optically active valence electron. The LS coupling scheme was used in calculations. Their P-(D-)channel results correspond to our 1^o (2^e) values. Unfortunately, Ref. [2] contains a number of numerical mistakes in calculations of a_{E2} coefficient, hindering a comparison. For example, for the P-channel, using Eq. (39) of Ref. [2] and their numerical values we obtain, $a_{E2,P} = -0.96 \times 10^{-8}\,\mathrm{cm/kV}$ which is an order of magnitude smaller than the published value. Similarly for the D-channel, based on Eq. (46) and numerical values of Ref. [2], we find $a_{E2,D} = 0.1 \times 10^{-8}\mathrm{cm/kV}$, a factor of 20 smaller than the published value. We present a detailed comparison with (revised) values of Ref. [2] for the two symmetries of intermediate states (1^o and 2^e) in Table IV. Although of the same order of magnitude, the individual contributions differ by signs. The most probable reason for the disagreement is the sensitivity to particulars of the treatment of correlations. For example, in computation of the D-channel contribution, Ref. [2] neglected intermediate states of the of ${}^3D$ symmetry. Hg is a heavy atom, and according to our table III, omitting the triplet contributions would increase $a_{E2,D}$ by a factor of three. There is a remarkable cancellation between individual channels (Table IV): our final result becomes an order of magnitude smaller than the recomputed value ($a_{E2} = -0.87 \times 10^{-8}\mathrm{cm/kV}$) of Ref. [2].

TABLE III: Breakdown of contributions to the reduced sums for the E2 Stark-induced transition. All quantities are in atomic units.

	$S_i^{E2}\,(J_n^\pi = 1^o)$		
n_nJ_n	$\langle 6s^2\,{}^1S_0\|\|\mathbf{r}\|\|n_nJ_n\rangle$	$\langle n_nJ_n\|\|Q^{(E2)}\|\|6s6p\,{}^3P_1\rangle$	contribution
$6s6p\,{}^3P_1$	-0.503	7.949	22.29
$6s6p\,{}^1P_1$	-2.956	-4.535	-54.41
$6s7p\,{}^3P_1$	-0.037	-5.460	-0.63
$6s7p\,{}^1P_1$	0.674	-1.647	3.42
$6s8p\,{}^3P_1$	-0.005	1.839	0.03
$6s8p\,{}^1P_1$	-0.286	1.652	1.35
$6s9p\,{}^3P_1$	-0.063	-2.875	-0.50
$6s9p\,{}^1P_1$	0.314	-0.269	0.23
Sum(10)			-28.23
Dalgarno-Lewis, Sum(∞)			-25.60
	$S_f^{E2}\,(J_n^\pi = 2^e)$		
n_nJ_n	$\langle 6s6p\,{}^3P_1\|\|\mathbf{r}\|\|n_nJ_n\rangle$	$\langle n_nJ_n\|\|Q^{(E2)}\|\|6s^2\,{}^1S_0\rangle$	contribution
$6s6d\,{}^1D_2$	1.570	-6.963	75.18
$6s6d\,{}^3D_2$	2.360	3.248	-52.61
$6s7d\,{}^1D_2$	0.576	-6.153	20.67
$6s7d\,{}^3D_2$	1.354	1.661	-13.10
$6s8d\,{}^1D_2$	0.263	-2.857	4.10
$6s8d\,{}^3D_2$	-1.363	0.422	3.14
$6s9d\,{}^1D_2$	-0.284	3.654	5.47
$6s9d\,{}^3D_2$	-1.281	-0.549	-3.71
Sum(10)			39.14
Dalgarno-Lewis, Sum(∞)			44.97

Fortunately, while a_{E2} has a poor accuracy, it turns out to be much smaller than a_{M1}, which, as shown below, can be computed reliably. There are two reduced sums to evaluate, $S_i^{M1}\,(1^o)$ and $S_f^{M1}\,(1^e)$. Non-relativistically, the magnetic-dipole operator is diagonal in the radial quantum numbers. This means that the only substantial contributions arise in the sum $S_i^{M1}\,(1^o)$. Indeed, we

TABLE IV: Comparison of different multipolar contributions to the Stark-induced absorption coefficients a_{M1} and a_{E2} in 1/(kV/cm). The first column gives the character of the multipole, the second column lists values of Ref. [2], and the third column gives the results of our computation. The notation $a[b] = a \times 10^b$ is used.

Contribution	Ref. [2]	This work
E2, 1^o	$-9.6[-9]^a$	2.4[−9]
E2, 2^e	$1.0[-9]^a$	−3.2[−9]
E2, total	$-8.7[-9]^a$	−0.85[−9]
M1, 1^e	0	−0.13[−9]
M1, 1^o	7.8[−9]	8.9[−9]
M1, total	7.8[−9]	8.86[−9]

[a]Values recomputed by us based on data of Ref. [2]; there are errors in numerical evaluations of Eq.(40), Eq.(46), and Eq.(47) of Ref. [2]. See text for details.

find from our fully relativistic analysis

$$S_i^{M1}(1^o) \approx 0.0285\,,$$
$$S_f^{M1}(1^e) \approx 0.0004\,.$$

The two dominant matrix elements entering $S_i^{M1}(1^o)$ are $\langle 6s6p\,^3P_1||Q^{(M1)}||6s6p\,^3P_1\rangle$ and $\langle 6s6p\,^3P_1||Q^{(M1)}||6s6p\,^1P_1\rangle$. Both matrix elements may be estimated non-relativistically (e.g., one could use the Landé formula for the first matrix element). Further, the term involving the $|6s6p\,^3P_1\rangle$ state is larger by roughly a factor of 5 than the contribution from the singlet state. As a result, the uncertainty in evaluating $S_i^{M1}(1^o)$ comes from the dipole matrix element entering this contribution, the already discussed $\langle 6s6p\,^3P_1||\mathbf{r}||6s^2\,^1S_0\rangle$. Incidentally, this is the very same matrix element that normalizes the absorption coefficient, so it cancels out in a_{M1}. Therefore, with about 25% accuracy

$$a_{M1} \approx \sqrt{\frac{2}{3}}\frac{\langle {}^3P_1||Q^{(M1)}||{}^3P_1\rangle}{E_f - E_i} \approx 1.19\times 10^{-8}/(\mathrm{kV/cm})\,,$$

where we used the non-relativistic value $\langle {}^3P_1||Q^{(M1)}||{}^3P_1\rangle = \left(\frac{3}{4}\right)\sqrt{6}\alpha$. Our full-scale Dalgarno-Lewis relativistic CI+MBPT calculation results in

$$a_{M1} = 0.886\times 10^{-8}/(\mathrm{kV/cm})\,,$$

and is consistent with the non-relativistic estimate. From the preceding discussion, it is clear that our theoretical value is stable with respect to neglected many-body corrections. Ref. [2] arrived at the result $a_{M1} = 0.780\times 10^{-8}/(\mathrm{kV/cm})$. This differs by 12% from our estimates.

Finally, we combine the contributions of the M1 and E2 interferences. We note that the poorly known E2 contribution is fortunately suppressed by a factor of 10 compared to the M1 coefficient. We find

$$a_{M1} + a_{E2} = 0.80\times 10^{-8}/(\mathrm{kV/cm})\,.$$

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